

REMARKS

Claims 1-12 are pending in the above-identified application.

Support for the changed to claim 1 is found at page 6, lines 1-15 of the specification. Support for the change to claim 10, as well as new claims 11 and 12 is found in Examples 1-5 described at pages 10-17 of the specification.

Request for Consideration of Information Disclosure Statement

It is requested that the Patent Examiner issue and initialed PTO-1449 form confirming the consideration and entry into the record of the documents cited in the Information Disclosure Statement filed November 14, 2003, pursuant to MPEP 609.

Issue Under 35 U.S.C. §112

Claim 1 has been rejected under 35 U.S.C. §112, second paragraph, as allegedly being indefinite, because this claim fails to specifically mention the function of the outer concentric tubes of the burner. Claim 1 has been amended so as to recite that gas may be fed to the outer concentric tubes of the burner. It is submitted that claim 1 complies with all applicable requirements under 35 U.S.C. §112 such that the above-noted rejection should be withdrawn.

Issues Under 35 U.S.C. §102(b) and 103(a)

Claim 10 has been rejected under 35 U.S.C. §102(b) or, in the alternative, under 35 U.S.C. §103(a) as being unpatentable over Rohr '560 (USP 5,340,560).

Claims 1-10 have been rejected under 35 U.S.C. §103(a) as being unpatentable over Rohr '560, optionally further in view of Nishimine '860 (USP 5,855,860).

The above-noted rejections are traversed for the following reasons.

Present Invention and Its Advantages

The present invention provides a novel and improved method for preparing a particulate silica and particulate silica thus prepared.

In accordance with the present invention, the method for preparing particulate silica involves premixing a gaseous organohalosilane with a flammable gas capable of generating water vapor when burned and a free oxygen-containing gas, and feeding the premix through a burner to a reaction chamber wherein the organohalosilane is subjected to flame hydrolysis. It is essential that the feed amount of the flammable gas capable of generating water vapor when burned, the configuration of the burner, and the

input rate of the gas premix ejected from the burner meet specific requirements as defined above.

When these requirements are met during the preparation of particulate silica using the organohalosilane as a raw material, the particulate silica thus obtained has a specific surface areas of 205 to 400 m<sup>2</sup>/g and a narrow particle size distribution of primary particles.

More specifically, the present invention provides the method for preparing particulate silica by feeding a mixture of a gaseous organohalosilane such as a methylchlorosilane, typically methyltrichlorosilane which is a by-product in the synthesis of dimethyldichlorosilane from metallic silicon and methyl chloride, with a flammable gas capable of generating water vapor when burned, typically hydrogen and a free oxygen-containing gas, typically air, to a reaction chamber through a burner, thereby subjecting the organohalosilane to flame hydrolysis and then oxidation reaction.

It has been found that when the amount of the flammable gas fed is ½ to 9 mol per mol of said organohalosilane and such that the amount of water vapor resulting from combustion of the flammable gas is 1 to 6 times the stoichiometric amount in reaction scheme (I) shown above, the burner used is one having a plurality of concentric tubes, and the gas mixture is fed to the center tube

of the burner such that it may have a linear velocity at the outlet of the center tube of 50 to 120 m/sec, calculated in the standard state, there is obtained particulate silica having a specific surface area of 205 to 400 m<sup>2</sup>/g and a narrow particle size distribution as demonstrated by a logarithmic standard deviation of primary particle diameter of up to 0.5.

The thus obtained particulate silica is suitable for a filler because silicone moldings filled therewith are highly transparent. This significant advantageous property is evident from the comparative test results summarized in Table 1 at page 17 of the present specification, which show that Examples 1-5 (present invention) exhibit advantageously enhanced transmittance over Comparative Examples 1 and 2 outside the scope of the present invention.

#### Distinctions Between Present Invention and Rohr '560

Rohr '560 may disclose the fumed silica aggregate having a surface area in the range of about 75 to 500 m<sup>2</sup>/g, and preferably 125 to 500 m<sup>2</sup>/g is obtained by the above method. However, these are only desired ranges of the surface area and there is no disclosure of any specific actual examples of the fumed silica with such a high surface area.

Additionally, the Office Action alleges on page 3 that "Rohr discloses a fumed silica having a surface area of  $132 \text{ m}^2/\text{g}$  and a standard deviation of 0.25 and a second fumed silica with surface area of  $154 \text{ m}^2/\text{g}$  and a standard deviation of 0.43".

However, this is incorrect. Rohr '560 discloses in column 3, lines 55 to 65 that "In addition, the flow rate and the composition of the feed mixture was altered to produce a larger aggregate fumed silica, Sample (B). The convex perimeter of Sample (A), made in accordance with the invention, was compared to fumed silica which was made in the same apparatus using a feed mixture outside the scope of the invention, Sample (B) and a commercial fumed silica "Cabot" M5 made by the Cabot Company of Tuscola, IL." Therefore, the only example within the scope of the invention of Rohr '560 is Sample (A) having a surface area of  $132 \text{ m}^2/\text{g}$  and a standard deviation of 0.25.

Thus, Rohr '560 completely fails to disclose any actual examples of fumed silica having as high as  $200 \text{ m}^2/\text{g}$  or more of a specific surface area, much less, the particulate silica having a specific surface area of 205 to  $400 \text{ m}^2/\text{g}$  and a narrow particle size distribution of primary particle diameter of up to 0.5 which is obtained by the inventive method of feeding the specific amount of the flammable gas and using the specific burner wherein the gas

mixture is fed to the center tube of the burner such that it has the specific linear velocity at the outlet of the center tube as described above.

The present invention can provide particulate silica having a high specific surface area and a narrow particle size distribution of primary particle diameter and ensuring the transparency of silicone moldings filled therewith. Such particulate silica according to the invention could not have been produced by the prior art methods as disclosed in the cited references.

Rohr '560 fails to disclose or teach the inventive method, the silica particulate having a high surface area and a narrow particle size distribution obtained by the method according to the invention.

It is further alleged on page 4 in the Office Action that "From Table II, the mole ratio of the hydrogen, i.e. the flammable gas to the methyltrichlorosilane can vary from  $(3.59/6.65=)0.54$  to  $(22.9/0.76=)30.13$ . This range overlaps the claimed range of "1/2 to 9)"."

However, the Office Action misunderstands the invention of Rohr '560 as discussed above. The Office Action alleges based on the disclosure of Sample B shown in Table II wherein 6.65 mol% of methyltrichlorosilane is used to 3.59 mol% of hydrogen (i.e.

$\text{H}_2/\text{methyltrichlorosilane} = 0.54 \text{ mol/mol}$ ). This is a Comparative Example which is outside the scope of the invention of Rohr '560, as the standard deviation of the average particle diameter or particle size distribution is too large.

Rohr '560 discloses the only one Working Example of Sample A wherein 0.76 mol% of methyltrichlorosilane is used to 22.90 mol% of hydrogen (i.e.  $\text{H}_2/\text{methyltrichlorosilane} = 30.1 \text{ mol/mol}$ ), which fails to correspond to or suggest the inventive method of feeding the flammable gas in an amount of  $\frac{1}{2}$  to 9 mol per mol of the organohalosilane and the feature thereof.

In the present invention, to obtain particulate silica having a minimized variation of primary particle diameter, the amount of water vapor that takes in the hydrolysis of organohalosilane is necessary. The reason is presumed as follows. The enlargement of particles occurs because the water vapor resulting from combustion of hydrocarbon of the like which is the functional group on the organohalosilane is unlikely to contribute to hydrolysis of the organohalosilane on account of a slow combustion rate involved therein, and inversely stated, because water vapor necessary for hydrolysis is depleted in proximity to the burner outlet. It is thus advantageous to use hydrogen featuring a high combustion rate. The water vapor also contributes to impart active OH groups to the

silica surface. Additionally, the presence of the flammable gas assists in forming a stable flame.

Rohr '560 fails to disclose or suggest the above feature of the invention.

Furthermore, Rohr '560 may disclose in Example 1 that "An annulus guard flame was maintained by feeding hydrogen in a concentric ring around the inner burner tube."

However, Rohr '560 fails to disclose or suggest the inventive method of feeding the specific amount of the flammable gas and ejecting the gas premix at the outlet of the center tube at the specific rate of the liner velocity of 50 to 120 m/sec. As described on page 7 of the specification, this gas linear velocity defined in the present invention is by far greater than the known velocities used in the prior art synthesis of particulate silica from organohalosilanes. Presumably the increased gas linear velocity at the burner outlet provides an increased slope of velocity in the flow field, enhanced turbulence and a reduced vortex scale. The enhanced turbulence increases the flame propagation area and burning velocity. This promotes hydrolysis of the organohalosilane in the flame whereby silica having a narrow particle size distribution is synthesized. It has been found that synthesis under these restricted conditions yields silica having a



specific surface area of 205 to 400 m<sup>2</sup>/g and a narrow particle size distribution as demonstrated by a logarithmic standard deviation of primary particle diameter of up to 0.5.

In view of the above, it is submitted that significant patentable distinctions exist between the present invention and Rohr '560 such that both of the above-noted rejections under 35 U.S.C. §102(b) and 103(a) should be withdrawn.

Distinctions Between Present Invention and Nishimine '860

Nishimine '860 discloses a method for purifying fine particulate silica comprising the steps of:

continuously feeding halide-bearing fine particulate silica, air and steam through an upright column from its bottom toward its top for forming a fluidized bed within the column at a gas linear velocity of 1 to 10 cm/sec and a temperature of 250 °C to 400 °C, the volume ratio of steam to air being at least 0.5/1, whereby steam causes the halide to be eliminated from the fine particulate silica, and

removing the fine particulate silica from which the halide has been eliminated from the top of said column.

Although Nishimine '860 discloses the use of a triple or quadruple tube burner, Nishimine '860 fails to disclose or suggest

feeding the specific amount of the flammable gas and injecting the gas mixture from the outlet of the center tube at the specific linear velocity employed in the present invention.

Consequently, significant patentable distinctions exist between the present invention and Nishimine '860.

In addition to the above, it is submitted that, even if Rohr '560 and Nishimine '860 were hypothetically combined, the resulting hypothetical combined disclosure would still fail to provide a basis for a motivation to those skilled in the art to obtain the method of the present invention of preparing particulate silica by subjecting a gaseous organohalosilane to flame hydrolysis with a flammable gas capable of generating water vapor when burned and a free oxygen-containing gas, wherein the flammable gas is fed in an amount of  $\frac{1}{2}$  to 9 mol per mol of the organohalosilane and such that the amount of water vapor resulting from combustion of the flammable gas is 1 to 6 times the stoichiometric amount of scheme (I), the burner has the plurality of concentric tubes, and the gas mixture is fed to the center tube of the burner such that it has a linear velocity at the outlet of the center tube of 50 to 120 m/sec.

Consequently, even assuming that Rohr '560 and Nishimine '860 could hypothetically be combined together, the resulting

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hypothetical combined disclosure would fail to support the alleged basis for the above-noted rejection under 35 U.S.C. §103(a), such that this rejection should be withdrawn.

It is submitted for the reasons stated above that the present claims define patentable subject matter such that this application should now be placed condition for allowance.

If any questions arise regarding the above matters, please contact Applicant's representative, Andrew D. Meikle (Reg. No. 32,868), in the Washington Metropolitan Area at the phone number listed below.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. §§ 1.16 or 1.17; particularly, extension of time fees.

Respectfully submitted,

BIRCH, STEWART, KOLASCH & BIRCH, LLP

By 

Andrew D. Meikle, #32,868

ADM:gmh

P.O. Box 747  
Falls Church, VA 22040-0747  
(703) 205-8000